Rapid Isotopic Analysis of Uranium Particles by Laser Ablation MC-ICP-MS

Supplementary Materials

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Supplement 1 – Text

Supplementary text S1. Calculation of Mass Bias Correction Factor

Mass bias and ion counter gain corrections are made by replicate analyses of the CAS-53-500 reference glass prior to, during, and after sample analyzes. The clear advantage of using the CAS-53-500 glass for correcting mass bias is that it contains subequal amounts of ^{235}U and ^{238}U, which means both isotopes can be measured using Faraday detectors, which lowers associated uncertainties compared to natural U in which the ^{235}U content is relatively low (~0.0072). The mass bias factor (MBF) is calculated assuming fractionation follows the exponential law, using Equation 1:

$$MBF = \frac{ln\left[\frac{\left(^{235}U/^{238}U\right)_{STD\_true}}{\left(^{235}U/^{238}U\right)_{STD\_measured}}\right]}{ln\left[M^{^{235}U}/M^{^{238}U}\right]} \quad \text{Equation 1}$$

Where the measured and true isotope ratios are those of a reference standard and M is the mass of the isotope. The MBF can then be used to correct the isotopic composition of an unknown using Equation 2:

$$\frac{^{235}U}{^{238}U}_{SMP\_unknown} = \frac{^{235}U}{^{238}U}_{SMP\_measured} \times \left(M^{^{235}U}/M^{^{238}U}\right)^{MBF} \quad \text{Equation 2}$$

The MBF is not a constant value and can vary both between and within analytical sessions. Thus, an added benefit of using a glass standard to calculate the MBF is that it can be analyzed multiple times during each session to characterize temporal changes in the MBF and apply an interpolated correction to the unknowns. A linear correction factor is applied between mass bias standards with a conservative expansion of uncertainty to account for potential non-linear drift. All relevant sources of measurement and instrument calibration uncertainty were propagated into final U isotope ratios via the Monte Carlo method.

Supplementary text S2. Calculation of Ion Counter Gain Correction Factor

The ion counter gain factor ensures equivalency between signals generated by Faraday detectors and signals generated on individual ion counters. Because the response of ion counters degrade over time and each

individual ion counter performs differently, individual gain factors are required for each detector. The CAS-53-500 reference standard has well characterized $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ratios, providing robust correction factors for IC3 (^{234}U) and IC1B (^{236}U). Once the MBF has been calculated (Equation 2), it is used to calculate the corrected $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ratios, as shown below in Equation 3:

$$\frac{^{23x}U}{^{238}U}_{SMP\_unknown} = \frac{^{23x}U}{^{238}U}_{SMP\_measured} \times \left(M^{^{23x}U}/M^{^{238}U}\right)^{MBF} \quad \text{Equation 3}$$

Where ^{23x}U refers to the minor isotope measured by ion counter. Once the mass bias corrected $^{23x}U/^{238}U$ ratio has been calculated for CAS-53-500 the corrected ratio is compared to the known ratio and the proportional difference is the ion counter gain, as shown below for IC3 (^{234}U):

$$IC3\ gain\ factor = \left(\frac{^{234}U}{^{238}U}_{STD\_corrected} \Big/ \frac{^{234}U}{^{238}U}_{STD\_true}\right) \quad \text{Equation 4}$$

Shifts in the ion counter gain factors occur between and within analytical sessions. For this reason, the ion counter gain corrections are also interpolated for each unknown.

Online ion counter gain corrections cannot be performed for IC2 (^{235}U) because of the relatively high abundance of ^{235}U in CAS-53-500, meaning it has to be measured by Faraday detector. Instead, the IC2 gain factor was estimated by analyzing a low-level U solution (NBS 960) by MC-ICP-MS with ^{235}U on IC2. The average IC2 gain factor over several analytical sessions was used as a constant during the laser ablation analyses. Although this produced acceptable results, we recognize that this is not an ideal solution as the gain factor may shift significantly in between analytical sessions, particularly if these sessions take place weeks or months apart. To address this in future studies we propose that an alternative reference material with a depleted ^{235}U content is used as a secondary standard purely to calculate the IC2 gain.

Supplement 2 – Tables

Table S1 – Isotopic compositions of the 6 reference glasses analyzed in this study. Isotopic compositions taken from measured by solution MC-ICP-MS at LLNL[1,2], unless otherwise stated. Uncertainties are external precisions associated with replicate analyses at the 2σ level.

	$^{234}U/^{238}U$	2σ	$^{235}U/^{238}U$	2σ	$^{236}U/^{238}U$	2σ
CAS3-53-500	0.007995	0.000035	1.1240	0.0029	0.005639	0.000024
CAS3-94-500	0.1682	0.0018	15.91	0.16	0.05031	0.00054
SAC-53-50	0.007806	0.000020	1.0985	0.0022	0.005498	0.000013
SAC-53-500	0.008017	0.000013	1.1283	0.0013	0.0056467	0.0000082
NIST 610[25]	0.00000945	0.00000005	0.00239555	0.00000047	0.00004314	0.00000004

Table S2 – Isotopic composition of 5 reference glasses measured in situ by NanoSIMS and SIM-SSAMS. Uncertainties are external precision associated with replicate analyses at the 2σ level. Here, the RSDs are used as a first-order indictor of the isotopic heterogeneity within in each glass.

Sample	Technique	$^{234}U/^{238}U$	2σ	RSD	$^{235}U/^{238}U$	2σ	RSD	$^{236}U/^{238}U$	2σ	RSD
CAS-94-500	NanoSIMS	0.166603	0.022711	13.6%	16.19534	1.676335	10.4%	0.049957	0.005356	10.7%
	SIM-SSAMS	0.1686	0.006083	3.6%	16.06051	0.268602	1.7%	0.050714	0.002338	4.6%
CAS-53-500	NanoSIMS	0.008026	0.000504	6.3%	1.139124	0.025083	2.2%	0.005541	0.000655	11.8%
	SIM-SSAMS	0.007974	0.000123	1.5%	1.120241	0.004958	0.4%	0.005677	6.37E-05	1.1%
SAC-53-50	NanoSIMS	0.007734	0.002855	36.9%	1.078263	0.076112	7.1%	0.005974	0.002397	40.1%
	SIM-SSAMS	0.007894	0.001556	19.7%	1.089784	0.03103	2.8%	0.005504	0.000748	13.6%
SAC-53-500	NanoSIMS	not measured								
	SIM-SSAMS	0.00792	0.000257	3.2%	1.122397	0.021632	1.9%	0.005616	0.000419	7.5%

Table S3 – U isotope ratios in the U200 reference standard previously characterized by solution MC-ICP-MS[3] and the DU sample from SRNL that was characterized using SIMS[4]. Uncertainties are external precisions associated with replicate analyses at the 2σ level.

Standard ID	$^{234}U/^{238}U$	2σ	$^{235}U/^{238}U$	2σ	$^{236}U/^{238}U$	2σ
U200	0.0015661	0.0000021	0.25119	0.00025	0.0026549	0.0000019
SRNL-DU	0.0000068	0.0000035	0.00173	0.00010	0.0000807	0.0000086

Table S4 – Uranium isotopic composition of reference glasses measured by laser ablation MC-ICP-MS

Sample Name	^{235}U detector	^{234}U/^{238}U	CSU (1σ)	^{235}U/^{238}U	CSU (1σ)	^{236}U/^{238}U	CSU (1σ)
CAS-94-500	Faraday	0.05109	0.00015	16.0956	0.0243	0.17011	0.00049
CAS-94-500	Faraday	0.05102	0.00014	16.1096	0.0251	0.17016	0.00046
CAS-94-500	Faraday	0.05091	0.00017	16.0917	0.0263	0.17012	0.00048
CAS-94-500	Faraday	0.05089	0.00017	16.0908	0.0260	0.16999	0.00055
CAS-94-500	Faraday	0.05115	0.00017	16.1303	0.0258	0.17073	0.00052
CAS-94-500	Faraday	0.05029	0.00016	16.1011	0.0241	0.16843	0.00045
CAS-94-500	Faraday	0.05105	0.00013	16.1052	0.0239	0.17054	0.00041
CAS-94-500	Faraday	0.05101	0.00014	16.1101	0.0242	0.17019	0.00042
CAS-94-500	Faraday	0.05103	0.00014	16.1075	0.0255	0.17025	0.00042
CAS-94-500	Faraday	0.05097	0.00014	16.1149	0.0263	0.17025	0.00044
CAS-94-500	Faraday	0.05080	0.00016	16.0934	0.0233	0.17036	0.00051
CAS-94-500	Faraday	0.05075	0.00017	16.0990	0.0230	0.16987	0.00055
CAS-94-500	Faraday	0.05085	0.00016	16.0882	0.0225	0.17009	0.00054
CAS-94-500	Faraday	0.05086	0.00017	16.0985	0.0235	0.16984	0.00053
CAS-94-500	Faraday	0.05078	0.00016	16.0780	0.0244	0.17010	0.00054
SAC-53-500	Faraday	0.005665	0.000017	1.1259	0.0015	0.008005	0.000023
SAC-53-500	Faraday	0.005655	0.000015	1.1262	0.0015	0.007999	0.000022
SAC-53-500	Faraday	0.005653	0.000017	1.1262	0.0015	0.008006	0.000021
SAC-53-500	Faraday	0.005658	0.000018	1.1257	0.0015	0.007991	0.000024
SAC-53-500	Faraday	0.005665	0.000018	1.1262	0.0015	0.008013	0.000025
SAC-53-500	Faraday	0.005600	0.000017	1.1258	0.0015	0.007930	0.000022
SAC-53-500	Faraday	0.005661	0.000015	1.1261	0.0015	0.008010	0.000020
SAC-53-500	Faraday	0.005658	0.000014	1.1257	0.0015	0.007992	0.000019
SAC-53-500	Faraday	0.005670	0.000014	1.1259	0.0015	0.007994	0.000020
SAC-53-500	Faraday	0.005655	0.000015	1.1258	0.0015	0.007997	0.000020
SAC-53-500	Faraday	0.005642	0.000019	1.1260	0.0015	0.008017	0.000027
SAC-53-500	Faraday	0.005637	0.000021	1.1262	0.0015	0.007981	0.000029
SAC-53-500	Faraday	0.005657	0.000019	1.1263	0.0015	0.008014	0.000027
SAC-53-500	Faraday	0.005640	0.000019	1.1262	0.0015	0.007982	0.000027
SAC-53-500	Faraday	0.005663	0.000038	1.1260	0.0015	0.008001	0.000054
SAC-53-50	Faraday	0.005529	0.000023	1.1007	0.0017	0.007832	0.000029
SAC-53-50	Faraday	0.005544	0.000023	1.1008	0.0017	0.007819	0.000027
SAC-53-50	Faraday	0.005502	0.000025	1.1023	0.0018	0.007825	0.000030
SAC-53-50	Faraday	0.005521	0.000025	1.1003	0.0018	0.007842	0.000030
SAC-53-50	Faraday	0.005545	0.000025	1.1002	0.0018	0.007849	0.000031
SAC-53-50	Faraday	0.005502	0.000023	1.1003	0.0017	0.007781	0.000026
SAC-53-50	Faraday	0.005500	0.000023	1.0996	0.0018	0.007829	0.000027
SAC-53-50	Faraday	0.005508	0.000021	1.1002	0.0019	0.007843	0.000029

SAC-53-50	Faraday	0.005543	0.000023	1.0991	0.0017	0.007800	0.000025
SAC-53-50	Faraday	0.005538	0.000022	1.1010	0.0018	0.007831	0.000027
SAC-53-50	Faraday	0.005515	0.000022	1.1010	0.0017	0.007841	0.000031
SAC-53-50	Faraday	0.005492	0.000025	1.1003	0.0017	0.007799	0.000033
SAC-53-50	Faraday	0.005505	0.000023	1.1005	0.0016	0.007821	0.000031
SAC-53-50	Faraday	0.005513	0.000024	1.0993	0.0016	0.007818	0.000033
SAC-53-50	Faraday	0.005522	0.000024	1.1001	0.0017	0.007824	0.000032
NIST610	IC	0.00004456	0.00000034	0.0023541	0.0000058	0.00001068	0.00000012
NIST610	IC	0.00004405	0.00000034	0.0023548	0.0000056	0.00001046	0.00000014
NIST610	IC	0.00004443	0.00000035	0.0023561	0.0000056	0.00001051	0.00000015
NIST610	IC	0.00004474	0.00000033	0.0023527	0.0000059	0.00001043	0.00000014
NIST610	IC	0.00004448	0.00000038	0.0023532	0.0000062	0.00001063	0.00000016
NIST610	IC	0.00004489	0.00000031	0.0023557	0.0000057	0.00001104	0.00000013
NIST610	IC	0.00004478	0.00000032	0.0023549	0.0000055	0.00001084	0.00000013
NIST610	IC	0.00004504	0.00000030	0.0023537	0.0000058	0.00001096	0.00000012
NIST610	IC	0.00004419	0.00000034	0.0023537	0.0000054	0.00001079	0.00000012
NIST610	IC	0.00004465	0.00000034	0.0023562	0.0000055	0.00001083	0.00000014
NIST610	IC	0.00004446	0.00000029	0.0023522	0.0000066	0.00001091	0.00000012
NIST610	IC	0.00004452	0.00000031	0.0023566	0.0000075	0.00001060	0.00000012
NIST610	IC	0.00004426	0.00000028	0.0023550	0.0000068	0.00001089	0.00000013
NIST610	IC	0.00004442	0.00000029	0.0023513	0.0000077	0.00001061	0.00000012
NIST610	IC	0.00004474	0.00000031	0.0023560	0.0000083	0.00001063	0.00000013

Note - Uncertainties are total internal precision associated with each individual measurement and the combined standard uncertainty (CSU) is provided at the 1σ level.

Table S5 – Uranium isotopic composition of CRM U200 particles measured by laser ablation MC-ICP-MS

Sample Name	^{235}U detector	^{234}U/^{238}U	CSU (1σ)	^{235}U/^{238}U	CSU (1σ)	^{236}U/^{238}U	CSU (1σ)
U200	Faraday	0.001555	0.000013	0.2505	0.0044	0.002559	0.000021
U200	Faraday	0.001569	0.000015	0.2504	0.0053	0.002546	0.000028
U200	Faraday	0.001545	0.000012	0.2515	0.0015	0.002486	0.000031
U200	Faraday	0.001581	0.000019	0.2532	0.0050	0.002515	0.000033
U200	Faraday	0.001443	0.000010	0.2510	0.0029	0.002325	0.000020
U200	Faraday	0.001558	0.000030	0.2519	0.0130	0.002572	0.000058
U200	Faraday	0.001574	0.000013	0.2533	0.0030	0.002535	0.000019
U200	Faraday	0.001095	0.000009	0.2513	0.0013	0.001675	0.000011
U200	Faraday	0.001527	0.000016	0.2509	0.0035	0.002517	0.000024
U200	Faraday	0.001530	0.000010	0.2516	0.0020	0.002501	0.000015
U200	Faraday	0.001568	0.000025	0.2569	0.0053	0.002673	0.000038
U200	Faraday	0.001516	0.000011	0.2532	0.0021	0.002497	0.000016
U200	Faraday	0.001547	0.000008	0.2508	0.0014	0.002530	0.000012
U200	Faraday	0.001544	0.000011	0.2524	0.0032	0.002552	0.000017
U200	Faraday	0.001453	0.000010	0.2517	0.0015	0.002343	0.000014
U200	Faraday	0.001604	0.000057	0.2536	0.0167	0.002396	0.000079
U200	Faraday	0.001560	0.000009	0.2510	0.0015	0.002481	0.000012
U200	Faraday	0.001570	0.000011	0.2506	0.0019	0.002540	0.000015
U200	Faraday	0.001579	0.000022	0.2515	0.0056	0.002469	0.000079
U200	Faraday	0.001596	0.000013	0.2507	0.0027	0.002531	0.000019
U200	Faraday	0.001465	0.000039	0.2542	0.0141	0.002579	0.000148
U200	Faraday	0.001553	0.000011	0.2526	0.0036	0.002544	0.000018
U200	Faraday	0.000403	0.000004	0.2597	0.0016	0.000669	0.000006
U200	Faraday	0.001508	0.000032	0.2508	0.0121	0.002536	0.000058
U200	Faraday	0.001583	0.000010	0.2511	0.0026	0.002594	0.000015
U200	Faraday	0.001577	0.000022	0.2506	0.0073	0.002530	0.000037
U200	Faraday	0.001473	0.000048	0.2475	0.0172	0.002507	0.000091
U200	Faraday	0.001536	0.000009	0.2494	0.0022	0.002602	0.000014
U200	Faraday	0.001553	0.000016	0.2506	0.0047	0.002597	0.000029
U200	Faraday	0.001525	0.000017	0.2505	0.0056	0.002558	0.000036
U200	Faraday	0.001553	0.000029	0.2464	0.0093	0.002558	0.000052
U200	Faraday	0.001415	0.000011	0.2516	0.0033	0.002275	0.000018
U200	Faraday	0.001324	0.000022	0.2476	0.0088	0.002093	0.000036
U200	Faraday	0.001522	0.000006	0.2515	0.0014	0.002541	0.000009
U200	Faraday	0.001562	0.000011	0.2511	0.0029	0.002627	0.000020
U200	Faraday	0.001587	0.000027	0.2488	0.0098	0.002600	0.000044
U200	Faraday	0.001307	0.000011	0.2506	0.0042	0.002141	0.000018
U200	Faraday	0.001563	0.000005	0.2516	0.0010	0.002610	0.000009
U200	Faraday	0.001584	0.000017	0.2501	0.0051	0.002614	0.000028
U200	Faraday	0.001554	0.000009	0.2508	0.0023	0.002615	0.000013

U200	Faraday	0.001544	0.000009	0.2524	0.0021	0.002620	0.000014
U200	Faraday	0.001549	0.000008	0.2509	0.0018	0.002563	0.000011
U200	Faraday	0.001543	0.000012	0.2515	0.0054	0.002567	0.000020
U200	Faraday	0.001571	0.000009	0.2509	0.0022	0.002580	0.000014
U200	Faraday	0.001535	0.000007	0.2514	0.0012	0.002593	0.000010
U200	Faraday	0.001563	0.000006	0.2510	0.0012	0.002562	0.000010
U200	Faraday	0.001551	0.000004	0.2512	0.0007	0.002562	0.000007
U200	Faraday	0.000314	0.000003	0.2675	0.0013	0.000504	0.000005
U200	Faraday	0.001471	0.000006	0.2509	0.0015	0.002430	0.000009

Note - Uncertainties are total internal precision associated with each individual measurement and the combined standard uncertainty (CSU) is provided at the 1σ level.

Table S6 – Uranium isotopic composition of SRNL-DU particles measured by laser ablation MC-ICP-MS

Sample Name	^{235}U detector	^{234}U/^{238}U	CSU (1σ)	^{235}U/^{238}U	CSU (1σ)	^{236}U/^{238}U	CSU (1σ)
SRNL DU	Faraday	0.0000073	0.0000006	0.0013329	0.0013809	0.0000806	0.0000015
SRNL DU	Faraday	0.0000060	0.0000005	0.0016321	0.0007527	0.0000819	0.0000014
SRNL DU	Faraday	0.0000075	0.0000004	0.0018222	0.0009097	0.0000803	0.0000014
SRNL DU	Faraday	0.0015190	0.0004098	0.1632718	0.2032309	0.0007227	0.0002846
SRNL DU	Faraday	0.0000074	0.0000005	0.0016282	0.0013420	0.0000764	0.0000015
SRNL DU	Faraday	0.0000077	0.0000005	0.0015331	0.0014296	0.0000781	0.0000016
SRNL DU	Faraday	0.0000055	0.0000006	0.0009771	0.0023713	0.0000794	0.0000022
SRNL DU	Faraday	0.0000078	0.0000015	NaN	NA	0.0000720	0.0000034
SRNL DU	Faraday	0.0000090	0.0000009	0.0014834	0.0017926	0.0000780	0.0000021
SRNL DU	Faraday	0.0000078	0.0000008	0.0016158	0.0024580	0.0000789	0.0000022
SRNL DU	Faraday	0.0000052	0.0000008	0.0007199	0.0033940	0.0000778	0.0000029
SRNL DU	Faraday	0.0000082	0.0000009	0.0017834	0.0026513	0.0000870	0.0000022
SRNL DU	Faraday	0.0000071	0.0000004	0.0017326	0.0010512	0.0000780	0.0000012
SRNL DU	Faraday	0.0000073	0.0000004	0.0016480	0.0010985	0.0000777	0.0000013
SRNL DU	Faraday	0.0000068	0.0000006	0.0017019	0.0011998	0.0000798	0.0000013
SRNL DU	Faraday	0.0000083	0.0000005	0.0015850	0.0014420	0.0000760	0.0000013
SRNL DU	Faraday	0.0000066	0.0000006	0.0008201	0.0017361	0.0000765	0.0000016
SRNL DU	Faraday	0.0000074	0.0000007	0.0017034	0.0014423	0.0000763	0.0000019
SRNL DU	Faraday	0.0000084	0.0000013	0.0016070	0.0160734	0.0000603	0.0000065
SRNL DU	Faraday	0.0000088	0.0000008	0.0005452	0.0041005	0.0000855	0.0000033
SRNL DU	Faraday	0.0000058	0.0000010	0.0011288	0.0055902	0.0000705	0.0000034
SRNL DU	Faraday	0.0000091	0.0000009	0.0039931	0.0038061	0.0000710	0.0000029
SRNL DU	Faraday	0.0000080	0.0000008	0.0010844	0.0041777	0.0000747	0.0000029
SRNL DU	Faraday	0.0000057	0.0000008	0.0005095	0.0042208	0.0000687	0.0000027
SRNL DU	Faraday	0.0000039	0.0000006	0.0002991	0.0044708	0.0000709	0.0000025
SRNL DU	Faraday	0.0000057	0.0000007	0.0018574	0.0040478	0.0000720	0.0000028
SRNL DU	Faraday	0.0000083	0.0000011	0.0036308	0.0069716	0.0000809	0.0000042
SRNL DU	Faraday	0.0000069	0.0000009	0.0015891	0.0041319	0.0000638	0.0000026
SRNL DU	Faraday	0.0000076	0.0000007	0.0005070	0.0041158	0.0000898	0.0000030
SRNL DU	Faraday	0.0000062	0.0000004	0.0013965	0.0009426	0.0000684	0.0000015
SRNL DU	Faraday	0.0000050	0.0000013	NaN	NA	0.0000905	0.0000050
SRNL DU	Faraday	0.0000075	0.0000008	0.0022959	0.0038077	0.0000773	0.0000028
SRNL DU	Faraday	0.0000075	0.0000007	0.0008754	0.0040699	0.0000875	0.0000030
SRNL DU	Faraday	0.0000076	0.0000016	0.0008941	0.0074027	0.0000723	0.0000039
SRNL DU	Faraday	0.0000038	0.0000009	0.0018312	0.0065452	0.0000896	0.0000035
SRNL DU	Faraday	0.0000069	0.0000005	0.0017689	0.0020076	0.0000751	0.0000017
SRNL DU	Faraday	0.0000065	0.0000008	0.0018296	0.0043395	0.0000771	0.0000031
SRNL DU	Faraday	0.0000102	0.0000011	0.0026755	0.0049377	0.0000654	0.0000034
SRNL DU	Faraday	0.0000074	0.0000005	0.0015519	0.0012775	0.0000682	0.0000016
SRNL DU	Faraday	0.0000076	0.0000008	0.0020864	0.0063121	0.0000696	0.0000037

SRNL DU	Faraday	0.0000084	0.0000014	0.0010274	0.0106334	0.0000743	0.0000039
SRNL DU	Faraday	0.0000058	0.0000005	0.0015301	0.0029002	0.0000817	0.0000021
SRNL DU	Faraday	0.0000070	0.0000002	0.0016049	0.0006379	0.0000787	0.0000009
SRNL DU	Faraday	0.0000068	0.0000004	0.0021469	0.0016867	0.0000811	0.0000017
SRNL DU	Faraday	0.0000068	0.0000004	0.0013130	0.0019647	0.0000733	0.0000017
SRNL DU	Faraday	0.0000066	0.0000004	0.0020031	0.0018684	0.0000810	0.0000017
SRNL DU	Faraday	0.0000088	0.0000006	0.0018375	0.0020997	0.0000834	0.0000022
SRNL DU	Faraday	0.0000070	0.0000005	0.0014349	0.0015817	0.0000806	0.0000017
SRNL DU	Faraday	0.0000091	0.0000005	0.0014335	0.0018715	0.0000805	0.0000018
SRNL DU	Faraday	0.0000080	0.0000006	0.0019243	0.0036440	0.0000907	0.0000027
SRNL DU	Faraday	0.0000059	0.0000004	0.0018042	0.0014279	0.0000821	0.0000015
SRNL DU	Faraday	0.0000074	0.0000005	0.0017950	0.0015658	0.0000779	0.0000016
SRNL DU	Faraday	0.0000078	0.0000005	0.0016157	0.0014485	0.0000829	0.0000017
SRNL DU	Faraday	0.0000043	0.0000005	0.0013723	0.0022444	0.0000523	0.0000019
SRNL DU	Faraday	0.0000098	0.0000003	0.0028200	0.0009598	0.0000971	0.0000012
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SRNL DU	Faraday	0.0000055	0.0000004	0.0010311	0.0017589	0.0000747	0.0000016
SRNL DU	Faraday	0.0000087	0.0000003	0.0020557	0.0005410	0.0000789	0.0000010
SRNL DU	Faraday	0.0000075	0.0000005	0.0013112	0.0021730	0.0000800	0.0000020
SRNL DU	Faraday	0.0000065	0.0000004	0.0017187	0.0015532	0.0000822	0.0000017
SRNL DU	Faraday	0.0000070	0.0000004	0.0020293	0.0015304	0.0000771	0.0000014
SRNL DU	Faraday	0.0000055	0.0000005	0.0015862	0.0024357	0.0000773	0.0000023
SRNL DU	Faraday	0.0000064	0.0000004	0.0016364	0.0016502	0.0000769	0.0000016
SRNL DU	Faraday	0.0000068	0.0000005	0.0013516	0.0020959	0.0000776	0.0000018
SRNL DU	Faraday	0.0000071	0.0000005	0.0012404	0.0016551	0.0000773	0.0000019
SRNL DU	IC	0.0000067	0.0000005	0.0016782	0.0000079	0.0000791	0.0000014
SRNL DU	IC	0.0000076	0.0000005	0.0016813	0.0000089	0.0000780	0.0000016
SRNL DU	IC	0.0000071	0.0000006	0.0016550	0.0000062	0.0000790	0.0000014
SRNL DU	IC	0.0000063	0.0000005	0.0016861	0.0000068	0.0000845	0.0000015
SRNL DU	IC	0.0000075	0.0000005	0.0016989	0.0000068	0.0000779	0.0000014
SRNL DU	IC	0.0000082	0.0000007	0.0016843	0.0000087	0.0000780	0.0000020
SRNL DU	IC	0.0000070	0.0000003	0.0016754	0.0000060	0.0000765	0.0000011
SRNL DU	IC	0.0000071	0.0000005	0.0016441	0.0000074	0.0000761	0.0000016
SRNL DU	IC	0.0000065	0.0000003	0.0016806	0.0000068	0.0000796	0.0000010
SRNL DU	IC	0.0000077	0.0000003	0.0016520	0.0000069	0.0000790	0.0000012
SRNL DU	IC	0.0000074	0.0000004	0.0016769	0.0000061	0.0000807	0.0000013
SRNL DU	IC	0.0000072	0.0000003	0.0016917	0.0000060	0.0000822	0.0000012
SRNL DU	IC	0.0000101	0.0000007	0.0009319	0.0000050	0.0000922	0.0000015
SRNL DU	IC	0.0000080	0.0000007	0.0016605	0.0000102	0.0000849	0.0000019
SRNL DU	IC	0.0000096	0.0000005	0.0016815	0.0000099	0.0000801	0.0000019
SRNL DU	IC	0.0000078	0.0000005	0.0016809	0.0000093	0.0000835	0.0000014
SRNL DU	IC	0.0000080	0.0000005	0.0016846	0.0000096	0.0000821	0.0000019

SRNL DU	IC	0.0000088	0.0000005	0.0016611	0.0000068	0.0000763	0.0000014
SRNL DU	IC	0.0000032	0.0000007	0.0016833	0.0000240	0.0000914	0.0000043
SRNL DU	IC	0.0000072	0.0000006	0.0016855	0.0000099	0.0000817	0.0000021
SRNL DU	IC	0.0000085	0.0000006	0.0017044	0.0000101	0.0000863	0.0000026
SRNL DU	IC	0.0000069	0.0000007	0.0017007	0.0000118	0.0000786	0.0000031
SRNL DU	IC	0.0000072	0.0000006	0.0016894	0.0000163	0.0000736	0.0000023
SRNL DU	IC	0.0000077	0.0000007	0.0016731	0.0000147	0.0000853	0.0000021
SRNL DU	IC	0.0000066	0.0000004	0.0003513	0.0000033	0.0000628	0.0000013
SRNL DU	IC	0.0000069	0.0000005	0.0016702	0.0000109	0.0000824	0.0000021
SRNL DU	IC	0.0000071	0.0000006	0.0016637	0.0000093	0.0000782	0.0000020
SRNL DU	IC	0.0000070	0.0000005	0.0016656	0.0000085	0.0000778	0.0000019
SRNL DU	IC	0.0000063	0.0000003	0.0004256	0.0000046	0.0000641	0.0000012
SRNL DU	IC	0.0000078	0.0000005	0.0016726	0.0000073	0.0000828	0.0000016
SRNL DU	IC	0.0000069	0.0000004	0.0016496	0.0000079	0.0000840	0.0000018
SRNL DU	IC	0.0000065	0.0000005	0.0016642	0.0000081	0.0000829	0.0000018
SRNL DU	IC	0.0000070	0.0000006	0.0016616	0.0000087	0.0000818	0.0000016
SRNL DU	IC	0.0000075	0.0000005	0.0016756	0.0000171	0.0000799	0.0000024
SRNL DU	IC	0.0000069	0.0000005	0.0016914	0.0000124	0.0000847	0.0000017
SRNL DU	IC	0.0000065	0.0000003	0.0004814	0.0000048	0.0000753	0.0000012
SRNL DU	IC	0.0000047	0.0000005	0.0016941	0.0000117	0.0000827	0.0000020
SRNL DU	IC	0.0000061	0.0000004	0.0016780	0.0000085	0.0000795	0.0000014
SRNL DU	IC	0.0000068	0.0000005	0.0016981	0.0000072	0.0000865	0.0000016
SRNL DU	IC	0.0000073	0.0000003	0.0005338	0.0000031	0.0000666	0.0000011
SRNL DU	IC	0.0000082	0.0000005	0.0016732	0.0000086	0.0000852	0.0000021

Note - Uncertainties are total internal precision associated with each individual measurement and the combined standard uncertainty (CSU) is provided at the 1σ level.

Supplement 3 - Figures

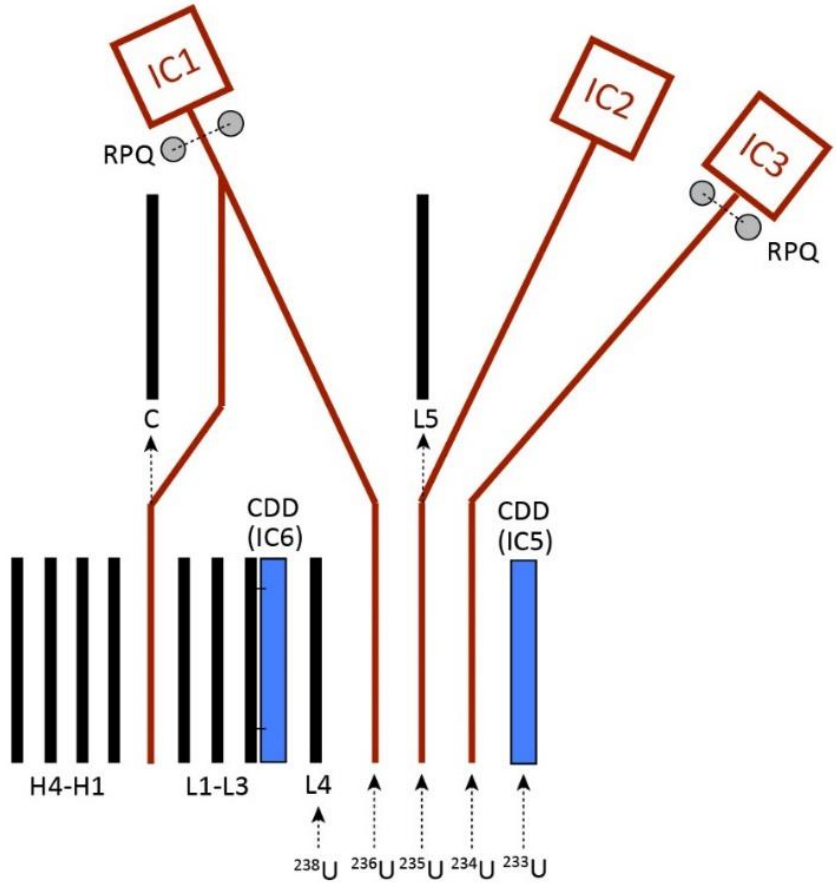

Fig. S1 – Detector array on the Neptune-Plus MC-ICP-MS. Detectors: Faradays (black), SEMs (red), and CDDS (blue). The IC1 and IC3 SEMs are fitted with energy filters (RPQs). U isotopes and associated detectors used for analysis are labeled.

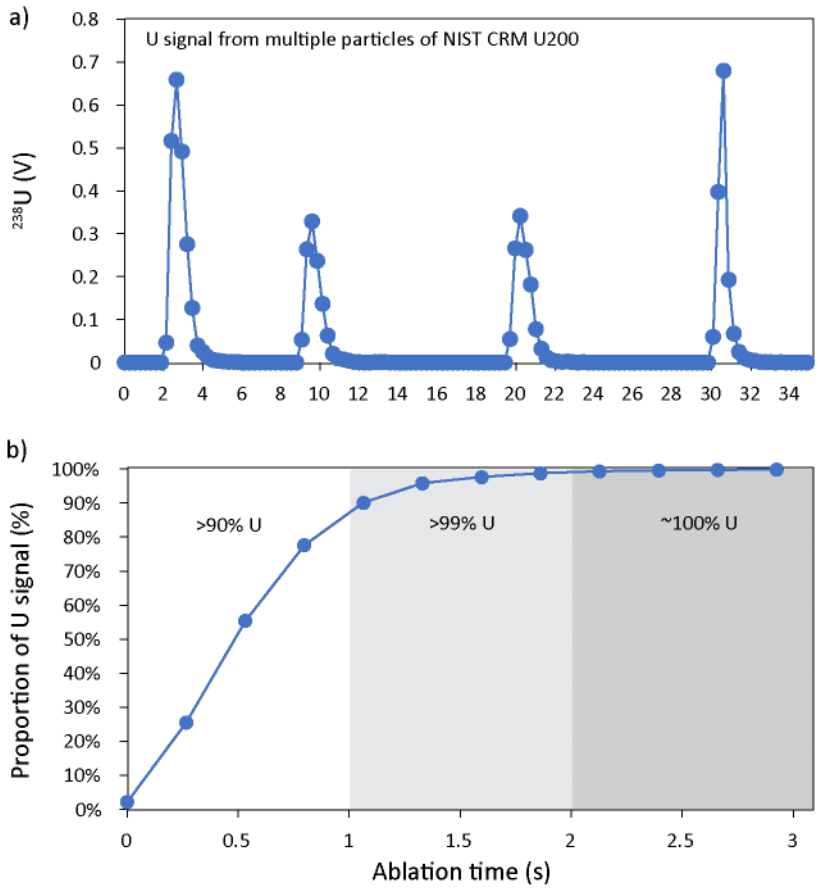

Fig. S2 – Laser ablation trace from rastering over a sample of U200 particles. Typical ^{238}U intensities generated from individual U200 particles are illustrated in a). The timeframe of U detection from a single particle is shown in horizontal axis of b). Note that the majority of U-signal is detected within 2 seconds of ablation initiation.

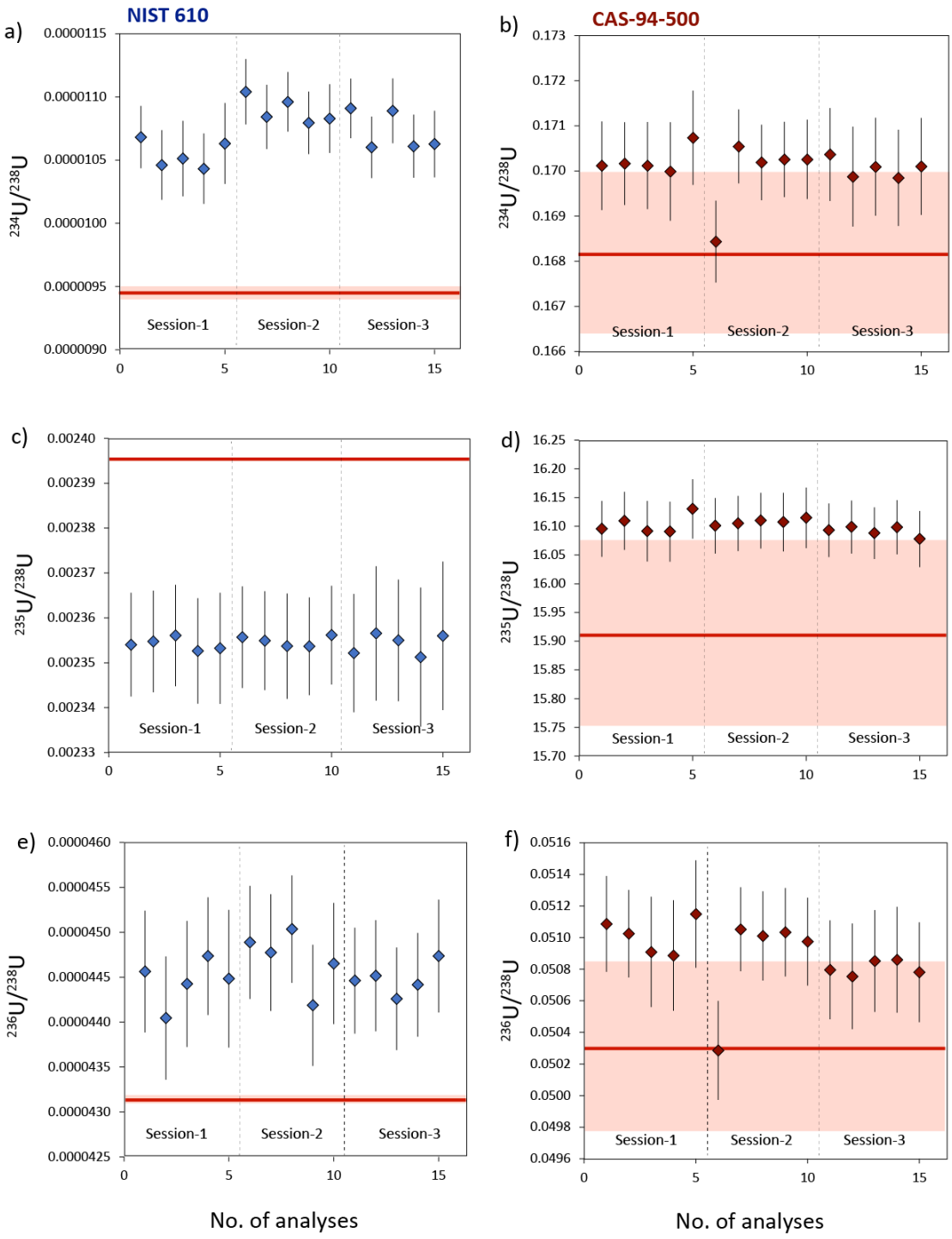

Fig. S3. Uranium isotope systematics in NIST 610 and CAS-94-500. Reference values and 2σ uncertainties are shown in red[1, 5]. Error bars on individual data points are 2σ and incorporate mass bias, ion counter gain and measurement uncertainties.

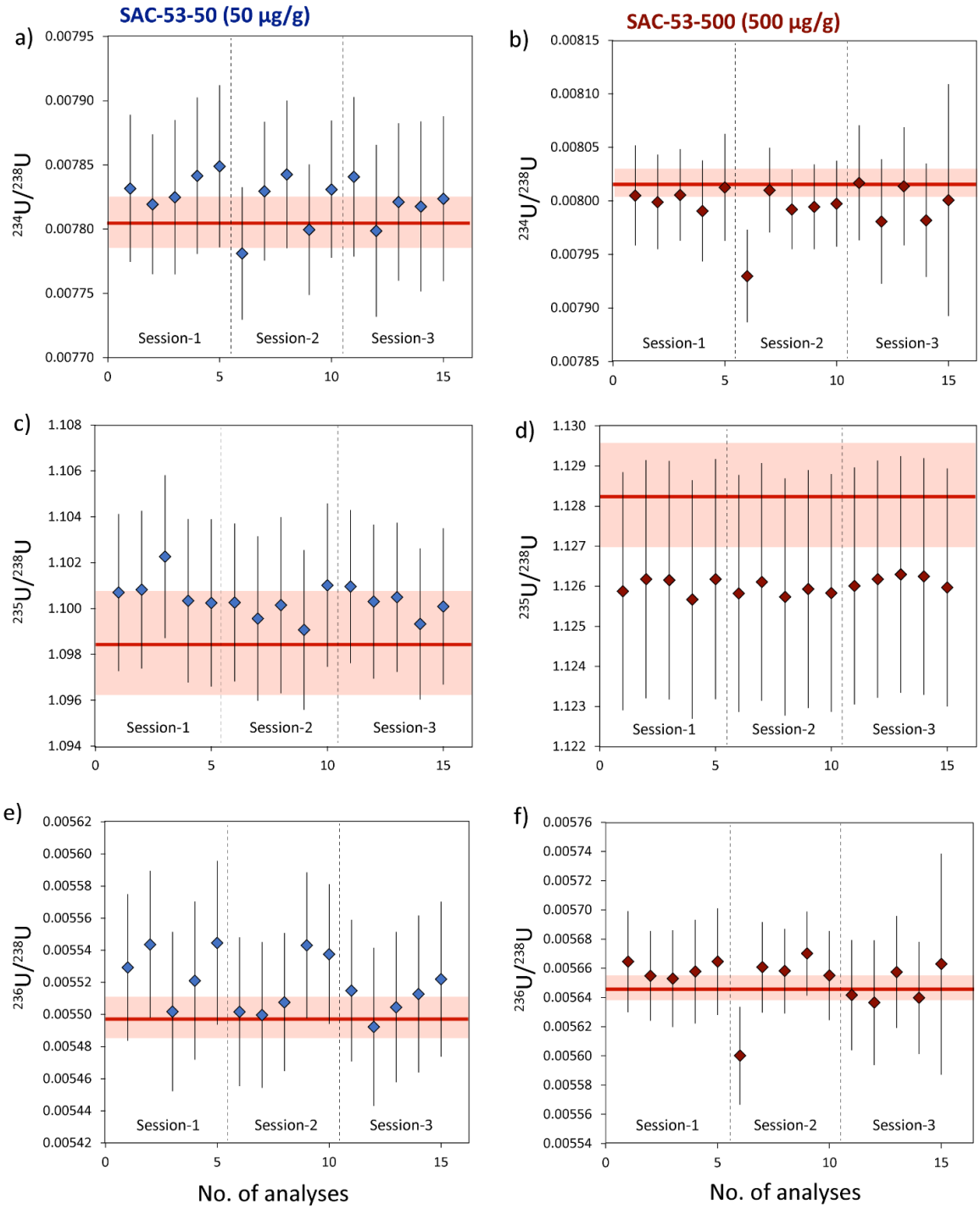

Fig. S4. Uranium isotope systematics in CAS-53-50 and CAS-53-500. Reference values and 2σ uncertainties are shown in red[1,5]. Error bars on individual data points are 2σ and incorporate mass bias, ion counter gain and measurement uncertainties.

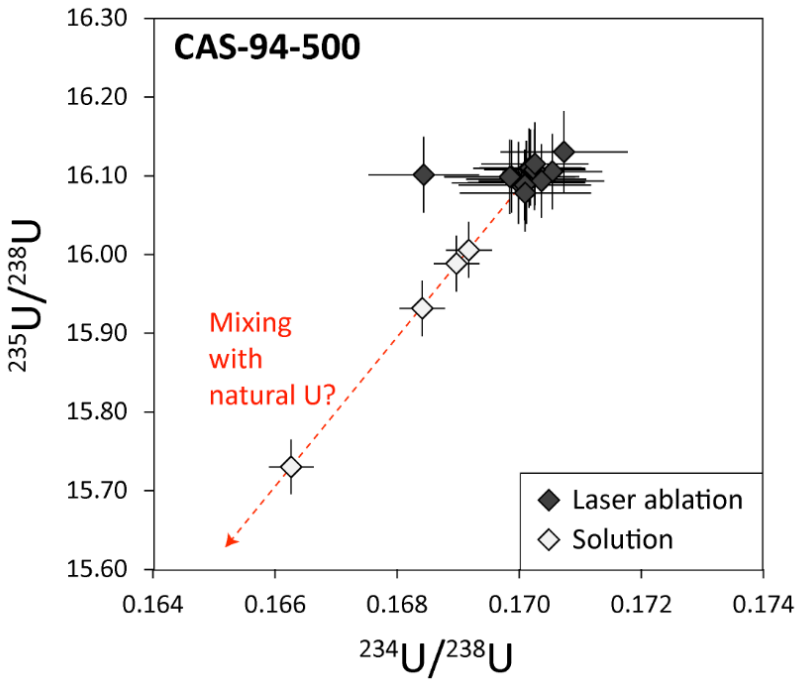

Fig. S5 – U isotope systematics in samples of CAS-94-500 measured by solution and LA-MC-ICP-MS. A clear mixing line extends from the laser ablation measurements to the solution analyses, indicating mixing between HEU and less enriched (i.e. natural) contaminant. Uncertainties are 2σ.

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